

# Magnetic Soret effect: application of the ferrofluid dynamics theory

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The ferrofluid dynamics theory is applied to thermodiffusive problems in magnetic fluids in the presence of magnetic fields. The analytical form for the magnetic part of the chemical potential and the most general expression of the mass flux are given. By employing these results to experiments, global Soret coefficients in agreement with measurements are determined. Also an estimate for a hitherto unknown transport coefficient is made.

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## I. INTRODUCTION

Magnetic fluids (MFs) are colloidal suspensions of ferromagnetic nanoparticles dispersed in a nonmagnetic carrier liquid. MFs behave superparamagnetically in a magnetic field and have a far reaching application potential spanning from sealants in rotary shaft to heat dissipaters in loud speaker coils [1] to carrier liquids for medical substances [2]. Starting in the mid-sixties of the last century, when MFs were first available, research on those fluids had been proceeding on the calm fairway of an established and well founded field of research. Particularly the theoretical work had been based on the achievements of the two pioneers, Rosensweig [3] and Shliomis [4, 5]. But 10 years ago a series of papers [6, 7, 8, 9] started to appear pointing specifically to the deficiencies of the microscopic approach in [4, 5] and proposing “... a general, strictly macroscopic approach relying solely on symmetry considerations, conservation laws, and thermodynamics.” [10]. This approach, called ferrofluid dynamics (FFD), sparked an impassioned discussion [10, 11] about which theory explains better the experimental facts for the reduced viscosity of a MF in an ac magnetic field [12] or for the magnetovortical resonance [13, 14]. That new theory also triggered an experiment [15] confirming a proposed nonzero transport coefficient which is zero in the microscopic approach [4, 5]. Other proposed effects as shear-excited sound [16, 17] await their confirmation yet.

For the description of thermal convection in magnetic fluid, the fluid has been considered as a one-component fluid with effective properties in many studies (see [18, 19, 20] and references therein). The limits of this coarse grained view onto the colloidal suspension of ferromagnetic nanoparticles are just being revealed. Considering a magnetic fluid as a binary liquid, the thermal convection is found to set in at Rayleigh numbers well below the threshold for a MF considered as a single-component fluid [21].

The thermodiffusive or Soret effect describes the establishment of concentration gradients in response to temperature gradients for a two-(or multi-)component fluid. Since the motion of the ferromagnetic nanoparticles in the MF can be influenced by external magnetic fields, the Soret effect in MFs shows a strong dependence on any nonzero magnetic field strength [22, 23, 24]. In a vertical layer the Soret coefficient  $S_T$  depends *nonmonotonously* on the strength of the field in the cases where the field is either parallel or perpendicular to the temperature gradient [22, 23]. Contrary, for both orientations of the magnetic field the Soret coefficient depends *monotonously* on the strength of the field if the layer is horizontal [24]. The changes of  $S_T$  can be up to six times its zero field value [23] and even a change of the sign of  $S_T$  was measured for strong fields [22, 23, 24].

The known theoretical approaches for the Soret effect in magnetic fluids [25, 26] need as an essential input an expression for the magnetophoretic velocity of the nanoparticles with respect to the carrier liquid. For that purpose certain microscopic properties are assumed as a dilute colloid containing spherical particles of equal size and the applicability of the Stokes hydrodynamic drag [25, 26]. Also assumptions about the deformation of the temperature distribution around the particle are made if its thermal conductivity is different from that of the surrounding carrier liquid [25]. A comparison with the known experimental results shows great differences: the microscopic theory [25, 27] gives only changes of  $S_T$  which are about *three* orders of magnitude smaller than the experimentally measured ones ([23] and Fig. 23 in [22]). Also in the frame of a thermodynamic approach [28] it is not possible to describe the drastic changes of  $S_T$  measured in the experiment. That means that with respect to thermodiffusive processes in MF in the presence of magnetic fields a wide gap between experiment and theory has to be bridged. Therefore it is the aim of this work to present a different approach, where in the frame of a macroscopic theory, the FFD, the experimental results can be described significantly better.

Usually an external temperature gradient causes both convection and thermodiffusion in any colloidal suspension. How these two effects are interacting with each other is not yet finally resolved as the discussion about the possibility of a state of relaxation-oscillation convection highlights [21, 29]. The mutual interference of convection and thermodiffusion is even more severe if additionally an external magnetic field is applied as in the case of MFs [22, 23, 24, 26, 30].

The problems caused by that mutual interference for the determination of the Soret effect are outlined in [24] and result in a new experimental setup for a horizontal layer of MF which is analyzed theoretically in this work.

## II. FERROFLUID DYNAMICS: CHEMICAL POTENTIAL AND MASS FLUX

The macroscopic FFD approach is presented without magnetodissipation, i.e. the magnetization  $\mathbf{M}$  is always parallel to the magnetic field  $\mathbf{H}$ , but with dissipative mass fluxes for the two constituents of the MF. The analysis will result in an analytical expression for the magnetic part of the chemical potential and a general expression for the mass flux without any assumption about the properties of the MF and the temperature distribution.

The principal structure of the ferrofluid dynamics theory was given in [9]. It is based firstly on general principles as symmetry considerations and conservations laws and on irreversible thermodynamics. The second independent component of which a macroscopic theory is made of is the set of material-dependent parameters as susceptibilities and transport coefficients. The latter can be determined by suitable experiments which are used here to determine transport coefficients for thermodiffusive processes in magnetic fluids in the presence of magnetic fields.

As usual in theories based on thermodynamical considerations, one starts with the thermodynamic energy density  $u$ . It is taken as a function of the entropy density  $s$ , the density  $\rho^{(1)}$  of the magnetic part of the fluid, the momentum density  $\mathbf{g} = \rho\mathbf{v}$ , the total density  $\rho$ , and the magnetic induction  $\mathbf{B} = \mu_0(\mathbf{M} + \mathbf{H})$  [9],

$$du = Tds + \tilde{\mu}_c d\rho^{(1)} + v_i dg_i + \mu^{(2)} d\rho + H_i dB_i, \quad (2.1)$$

where  $\tilde{\mu}_c = \tilde{\mu}^{(1)} - \tilde{\mu}^{(2)}$  is the difference in the chemical potentials of the two constituents. The conservation laws for the density of the magnetic and nonmagnetic part  $\rho^{(2)}$  are

$$\partial_t \rho^{(1)} = -\nabla_i \left( \rho^{(1)} v_i - j_i^D \right), \quad (2.2)$$

$$\partial_t \rho^{(2)} = -\nabla_i \left( \rho^{(2)} v_i + j_i^D \right), \quad (2.3)$$

where  $j_i^{D(1)} = -j_i^{D(2)} = j_i^D$  was used to ensure the conservation of the total density  $\rho = \rho^{(1)} + \rho^{(2)} = \phi \rho_m + (1 - \phi) \rho_{cl}$ . The density of the magnetic particles (carrier liquid) is denoted by  $\rho_m$  ( $\rho_{cl}$ ) and  $\phi$  is the volume fraction of magnetic particles in the fluid. The dissipative mass flux  $\mathbf{j}^D$  is proportional to the gradient of the chemical potential with  $\tilde{\mu}_c = \tilde{\mu}_c(\rho, \rho^{(1)}, T, \mathbf{v}, \mathbf{H})$  and the temperature gradient [31]. It is assumed that the magnetic part of the chemical potential can be separated [32],

$$\tilde{\mu}_c = \mu_c(\rho, \rho^{(1)}, T, \mathbf{v}) + \mu_c^m(\rho, \rho^{(1)}, T, \mathbf{v}, \mathbf{H}). \quad (2.4)$$

This assumption guarantees a nonzero chemical potential for  $\mathbf{H} = 0$  and is confirmed by calculations for MF with chains, where the magnetic part contributes additive to the total chemical potential [33]. The nonmagnetic part of the chemical potential is given by  $\mu_c = (k_B T / m_m) \ln c_1 - (k_B T / m_{cl}) \ln c_2$ , where  $m_m$  ( $m_{cl}$ ) is the mass of a magnetic (carrier liquid) particle [31].

The experiments [22, 23, 24] show that *any* nonzero strength of the magnetic field influences the thermodiffusive processes. Thus the general ansatz for the dissipative mass flux is (following the notation in [9])

$$j_i^D = \xi_1 \nabla_i T + \xi \nabla_i \tilde{\mu}_c + \xi_{\parallel} M_i M_j \nabla_j \tilde{\mu}_c + \xi_{\times} \varepsilon_{ijk} M_j \nabla_k \tilde{\mu}_c. \quad (2.5)$$

Whereas the first two terms characterize isotropic mass fluxes caused by gradients in the temperature or in the chemical potential, the last two terms describe anisotropic mass fluxes, namely parallel and perpendicular to the direction of  $\mathbf{M}$ . The last term corresponds to that one in the analogous ansatz for the heat flux, where the phenomenon is called transversal Righi-Leduc effect [31] since the primary current is perpendicular to the produced effect.

It was emphasized in [9] that the "proliferation of transport coefficients", i.e.  $\xi \rightarrow (\xi, \xi_{\parallel}, \xi_{\times})$ , takes place in the case of strong magnetic fields. But the experiments show that at least for thermodiffusive processes that general statement seems not to be true. In the figures in [22, 23, 24] with respect to the changes of  $S_T$  it is evident that small magnetic fields in the order of less than 50 kA/m are sufficient to generate effects, where one can clearly distinguish between a parallel or a perpendicular orientation between temperature gradient and field. Therefore the in [9] firstly introduced coefficients  $\xi_{\parallel}$  and  $\xi_{\times}$  are considered here as nonzero for all magnetic field strengths.

With the above given dependences of the chemical potential in Eq. (2.4), its gradient is

$$\nabla_i \tilde{\mu}_c = \frac{\partial \tilde{\mu}_c}{\partial \rho} \nabla_i \rho + \frac{\partial \tilde{\mu}_c}{\partial \rho^{(1)}} \nabla_i \rho^{(1)} + \frac{\partial \tilde{\mu}_c}{\partial T} \nabla_i T + \frac{\partial \tilde{\mu}_c}{\partial v_j} \nabla_i v_j + \frac{\partial \mu_c^m}{\partial H_j} \nabla_i H_j. \quad (2.6)$$

The first expression in Eq. (2.6) will become later the term for the barodiffusion and can be neglected in an incompressible fluid not subjected to any pressure gradient. For the fourth and fifth term [28, 34] hold

$$\frac{\partial \tilde{\mu}_c}{\partial v_j} = -\frac{\partial(\rho v_j)}{\partial \rho^{(1)}} \equiv 0, \quad (2.7)$$

$$\frac{\partial \mu_c^m}{\partial H_j} = -\mu_0 \frac{\partial}{\partial \rho^{(1)}} (H_j + M_j) = -\mu_0 \frac{\partial M_j}{\partial \rho^{(1)}}. \quad (2.8)$$

The transformation  $\tilde{u} = u - v_j g_j - H_j B_j$  was made in order to match the dependences of the energy density and the chemical potential and usage of the fact that derivatives of quantities are zero which are independent of each other. From the last equality the analytical result for the magnetic part of the chemical potential follows,

$$\mu_c^m = -\mu_0 \int_0^H \frac{\partial M}{\partial \rho^{(1)}} dH', \quad (2.9)$$

where  $M$  and  $H$  denote the absolute value of the magnetic field and the magnetization. Eq. (2.9) allows a direct determination of  $\mu_c^m$  if the magnetization  $M(H, \rho^{(1)}, T)$  is known without any assumption about the properties of the MF in contrast to [27, 28, 33, 34]. According to these references the determination of the chemical potential needs the knowledge of quantities like the volume concentration of the nanoparticles [27, 33] or the strength of the magnetodipole interaction [33] or the effective field experienced by a single particle in the MF [27, 28, 34]. Compared with the effort to evaluate these microscopic details, the advantage of the macroscopic approach of the FFD is apparent. A measurement of the magnetization as function of the magnetic field and the density is sufficient to determine the chemical potential for any magnetic fluid.

Inserting Eq. (2.6) into Eq. (2.5) and using Eqs. (2.7, 2.8) an expression for the mass flux results,

$$\begin{aligned} \frac{\mathbf{j}^D}{\rho} = & \left( \frac{\xi_1}{\rho} + \frac{\xi}{\rho} \frac{\partial \mu_c}{\partial T} \right) \nabla T + \frac{\xi}{\rho} \frac{\partial \mu_c^m}{\partial T} \nabla T + \frac{\partial \tilde{\mu}_c}{\partial T} \left[ \frac{\xi_{\parallel}}{\rho} \mathbf{M} (\mathbf{M} \nabla T) + \frac{\xi_{\times}}{\rho} (\mathbf{M} \times \nabla T) \right] \\ & + \xi \frac{\partial \mu_c}{\partial \rho^{(1)}} \nabla c_1 + \xi \frac{\partial \mu_c^m}{\partial \rho^{(1)}} \nabla c_1 + \frac{\partial \tilde{\mu}_c}{\partial \rho^{(1)}} [\xi_{\parallel} \mathbf{M} (\mathbf{M} \nabla c_1) + \xi_{\times} (\mathbf{M} \times \nabla c_1)] \\ & - \frac{\mu_0}{\rho} \frac{\partial M}{\partial \rho^{(1)}} [\xi \nabla H + \xi_{\parallel} \mathbf{M} (\mathbf{M} \nabla H) + \xi_{\times} (\mathbf{M} \times \nabla H)] , \end{aligned} \quad (2.10)$$

which is generally valid, independent of the size distribution of the magnetic particles, concentration inhomogeneities in the suspension or the form of the temperature gradient. Therefore Eq. (2.10) is the generalization of the mass flux given in [25]. The concentration of the magnetic particles  $c_1 = \rho^{(1)}/\rho$  is defined by means of the mass fraction of the total density  $\rho$  [31]. The first four terms describe mass flow caused by thermophoresis ( $\sim \nabla T$ ), the second four terms by diffusiohoresis ( $\sim \nabla c_1$ ), and the last three by magnetophoresis ( $\sim \nabla H$ ). There are two unknown transport coefficients,  $\xi_{\parallel}$  and  $\xi_{\times}$ , since for zero magnetic field, Eq. (2.10) reduces to the classical result (see Eq. (227), Chapt. XI in [31])

$$\begin{aligned} \frac{\mathbf{j}^D}{\rho} = & \left( \frac{\xi_1}{\rho} + \frac{\xi}{\rho} \frac{\partial \mu_c}{\partial T} \right) \nabla T + \xi \frac{\partial \mu_c}{\partial \rho^{(1)}} \nabla c_1 \\ = & c_1 c_2 D_T \nabla T + D_c \nabla c_1 \end{aligned} \quad (2.11)$$

with  $(D_T)$   $D_c$  the (thermal) diffusion coefficient known for MFs from previous experiments [35] and  $c_2 = 1 - c_1$ . According to the philosophy of the FFD approach, the determination of the unknown transport coefficients  $\xi_{\parallel}$  and  $\xi_{\times}$  needs suitable experiments which were conducted just recently [24].

### III. APPLICATION TO EXPERIMENTS AND DISCUSSION

According to the experiments for a horizontal layer of MF of thickness  $h$  [24], a horizontally unbounded layer of a dielectric, viscous, and incompressible MF sandwiched between two perfect conducting plates is considered. The lower plate is cooled to  $T_1$  and the upper one is heated to  $T_2$ . The resulting temperature gradient stabilizes the quiescent conductive state. From the equation of heat conduction,

$$\frac{\partial T}{\partial t} = \kappa \Delta T, \quad (3.1)$$

and the boundary conditions,

$$T(z = h/2) = T_2 \quad T(z = -h/2) = T_1 , \quad (3.2)$$

the temperature profile of the conductive state

$$T = T_0 + \frac{(T_2 - T_1)}{h} z \quad (3.3)$$

follows with  $T_0 = (T_1 + T_2)/2$  and  $\kappa$  denotes the thermal diffusivity. Since the plates are impenetrable, the diffusion equation,

$$\frac{\partial c_1}{\partial t} = \text{div} \left( \frac{\mathbf{j}^D}{\rho} \right) , \quad (3.4)$$

has to be supplemented with the boundary condition

$$j_z^D(z = \pm h/2) = 0. \quad (3.5)$$

Rearranging this boundary condition with the help of Eq. (2.11),

$$\frac{-h}{c_1 c_2 (T_2 - T_1)} \frac{\partial c_1}{\partial z} \Big|_{z=\pm h/2} = \frac{D_T}{D_c} = S_T , \quad (3.6)$$

the Soret coefficient in the zero field case can be expressed. In the same way the global Soret coefficient, measured in [24], in the presence of a magnetic field can be determined by using Eq. (2.10).

If a spatially homogeneous static magnetic field is applied perpendicular to the layer, the resulting magnetic field gradient inside the fluid is parallel to the temperature gradient. Therefore this setup is called parallel and is analyzed first.

Taking diffusion processes into account, the magnetization in the fluid can be written in the form

$$\mathbf{M} = \left[ M_0 + \chi(H - H_0) - K(T - T_0) + \frac{\partial M}{\partial \phi}(\phi - \phi_0) \right] \mathbf{e}_z , \quad (3.7)$$

where  $M_0 = M_0(H_0, T_0, \phi_0)$  is the reference magnetization belonging to the reference values  $H_0$ ,  $T_0$ , and  $\phi_0$  for the magnetic field, the temperature, and the volume fraction. Extending the expressions given in [18], magnetization and magnetic field for the conductive state are

$$M = M_0 + \frac{K(T_1 - T_2)}{h(1 + \chi)} z + N(c_1 - c_{1,0}) , \quad (3.8)$$

$$H = H_0 - \frac{K(T_1 - T_2)}{h(1 + \chi)} z - N(c_1 - c_{1,0}) , \quad (3.9)$$

with the susceptibility  $\chi = \partial M / \partial H$ , the pyromagnetic coefficient  $K = -\partial M / \partial T$ , the densomagnetic coefficient  $N = \partial M / \partial c_1 = (\rho / \rho_m)(\partial M / \partial \phi)$ , and  $c_{1,0} = c_1(T_0)$ . Inserting Eqs. (3.8, 3.9) into Eq. (2.10) and rearranging the boundary condition (3.5) in the same manner as in the zero field case, the global Soret coefficient in the parallel setup reads:

$$S_T^{\parallel} = \frac{S_T + \frac{1}{c_{1,0} c_{2,0} \rho} \left[ \frac{\xi}{D_c} \frac{\partial \mu_c^m}{\partial T} + \frac{\xi_{\parallel}}{D_c} \frac{\partial \mu_c}{\partial T} M^2 - \mu_0 \frac{\partial M}{\partial \rho^{(1)}} \left( \frac{\xi}{D_c} + \frac{\xi_{\parallel}}{D_c} M^2 \right) \frac{K}{(1 + \chi)} \right]}{1 + \frac{\xi}{D_c} \frac{\partial \mu_c^m}{\partial \rho^{(1)}} + \frac{\xi_{\parallel}}{D_c} \frac{\partial \mu_c}{\partial \rho^{(1)}} M^2 + \frac{\mu_0}{\rho} \frac{\partial M}{\partial \rho^{(1)}} \left( \frac{\xi}{D_c} + \frac{\xi_{\parallel}}{D_c} M^2 \right) N} . \quad (3.10)$$

Knowing  $M(H, \rho^{(1)}, T)$  in analytical form allows one to calculate  $\mu_c^m$  and its derivatives. The measured magnetization curve (from Fig. 57 in [22]) could be nicely fitted with  $M = M_b \lambda_{\phi} \phi L(\lambda_d \alpha)$ , where  $L(\lambda_d \alpha) = \coth(\lambda_d \alpha) - 1/(\lambda_d \alpha)$  is the Langevin function,  $\alpha = \mu_0 m H / (k_B T)$  the Langevin parameter,  $m = M_b \pi d^3 / 6$  the magnetic moment of a particle, and  $k_B$  the Boltzmann constant.  $\lambda_d$  and  $\lambda_{\phi}$  are two geometrical fit parameters. They reflect small deviations from the volume fraction  $\phi = 0.2$  and the  $\delta$ -shaped size distribution (Fig. 59 in [22]). Using  $\lambda_d = 0.99$ ,  $\lambda_{\phi} = 0.84$  and the material data  $M_b = 450$  kA/m (magnetization of the magnetic bulk solid),  $d = 9$  nm,  $\rho_m = 5.15$  g/cm<sup>3</sup> from [22], the solid line in Fig. 1 shows a very good agreement with the measured magnetization ( $\circ$ ). Considering the chosen

values for  $\lambda_d$  and  $\lambda_\phi$ , only the volume fraction  $\phi$  had to be adjusted to the measured data. Variations in  $\phi$  are likely caused by a nonmagnetic surface layer of the nanoparticles [36] and its solvability in the carrier liquid. According to the statement at the beginning of the paragraph, one has

$$\mu_c^m = \frac{\lambda_\phi}{\lambda_d} \frac{M_b k_B T}{2\rho_m m} \left\{ \ln [\coth^2(\lambda_d \alpha) - 1] + 2 \ln(\lambda_d \alpha) \right\}, \quad (3.11)$$

from which one can calculate the derivatives with respect to  $T$  and  $\rho^{(1)} = \phi \rho_m$ .

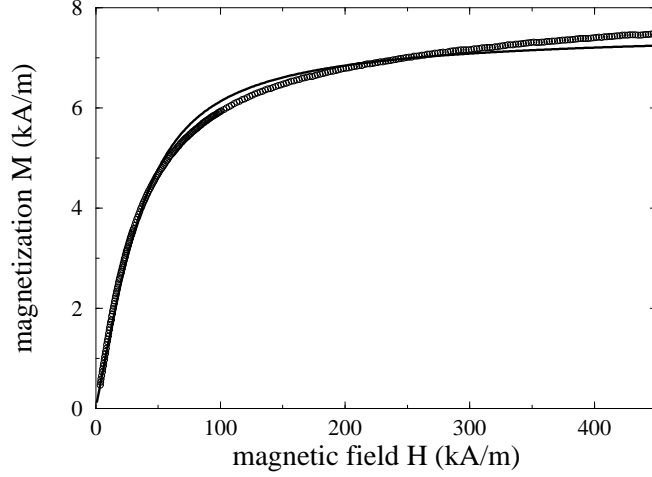


FIG. 1: Experimental data ( $\circ$ ) from Fig. 57 in [22] and theory (solid line) for the magnetization at room temperature  $T = 293$  K. The details of the used Langevin function are given in the text.

With the pyromagnetic coefficient  $K$  taken from Fig. 4 in [22], it remains the four unknowns  $S_T$ ,  $D_c$ ,  $\xi$ , and  $\xi_{\parallel}$  in Eq. (3.10). To fit  $S_T^{\parallel}$  to the experiment, the combined quantities  $\xi/D_c$  and  $\xi_{\parallel}/D_c$  are used as fit parameters, since  $S_T = 0.15 \text{ K}^{-1}$  was measured in the zero field case [24] but not  $D_c$ . The solid line in Fig. 2 gives the best two parameter fit, yielding  $\xi/D_c = 8.2 \text{ kg s}^2/\text{m}^5$  and  $\xi_{\parallel}/D_c = -1.41 \cdot 10^{-7} \text{ kg s}^2/(\text{m}^3 \text{ A}^2)$ . The difference in the absolute values of about 8 orders of magnitude is not surprisingly, since one would assume such a relation according to the argument that anisotropic fluxes in the mass flux (2.5) are relevant only for strong fields [9]. Inspecting Eq. (3.10) closer, it is revealed that  $\xi_{\parallel}/D_c$  is multiplied by  $M^2$  which gives already for small magnetic fields a factor of  $\sim 10^6$ . The two other terms are not so relevant because  $0 \leq \partial\mu_c/\partial T \leq 0.016 \text{ J}/(\text{K kg})$  and  $0 \leq \partial\mu_c/\partial\rho^{(1)} \leq 0.35 \text{ Jm}^3/\text{kg}^2$  for  $0 \leq H \leq 350 \text{ kA/m}$ . To underline the relevance of  $\xi_{\parallel}/D_c$  even for small fields, the dot-dashed line in Fig. 2 displays  $S_T^{\parallel}$  for  $\xi_{\parallel}/D_c = 0$  and all other parameters as before. Now the theoretical curve misses the measured data ( $\square$ ) clearly. Taking a typical value for the diffusion coefficient,  $D_c \sim 10^{-11} \text{ m}^2/\text{s}$  [35], the new transport coefficient can be estimated to  $\xi_{\parallel} \sim -10^{-18} \text{ kg s}/(\text{m A}^2)$  for the MF in [22, 24]. Thus those experiments deliver the necessary input for determining the material-dependent transport coefficients which are a priori unknown in a macroscopic theory as the FFD. Another example for the experimental determination of diffusion and thermodiffusion coefficients is presented in [35], whereas in [37] these coefficients were calculated on the basis of a microscopic theory.

In contrast to the parallel setup, in the perpendicular setup the spatially homogeneous static magnetic field is applied perpendicular to the temperature gradient, i.e. the magnetic field is parallel to the layer. The diffusion equation gets now the form

$$\frac{\partial c_1}{\partial t} = \frac{\partial \tilde{\mu}}{\partial \rho^{(1)}} \left[ (\xi + \xi_{\parallel} M^2) \frac{\partial^2 c_1}{\partial x^2} + \xi \left( \frac{\partial^2 c_1}{\partial y^2} + \frac{\partial^2 c_1}{\partial z^2} \right) \right]. \quad (3.12)$$

The boundary condition for the  $z$ -component of the mass flux yields

$$\frac{\partial c_1}{\partial z} = -\frac{\xi_{\perp} M}{\xi} \frac{\partial c_1}{\partial y} - \frac{c_1 c_2 D_T + \frac{\xi}{\rho} \frac{\partial \mu_c^m}{\partial T}}{D_c + \xi \frac{\partial \mu_c^m}{\partial \rho^{(1)}}} \text{ at } z = \pm h/2. \quad (3.13)$$

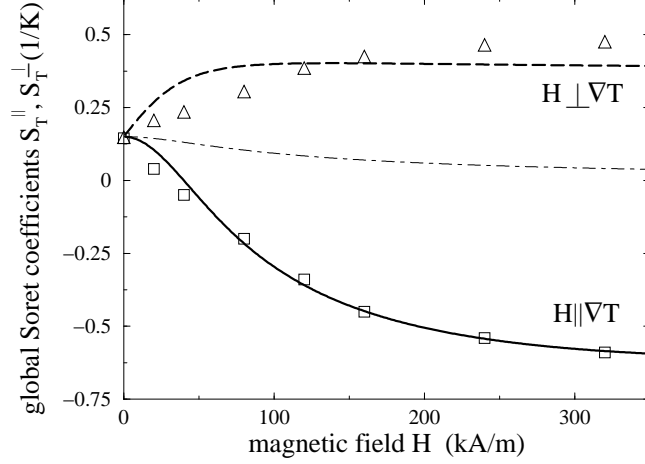


FIG. 2: Global Soret coefficients  $S_T^{\parallel}$  and  $S_T^{\perp}$  against the magnetic field strength for the parallel ( $H \parallel \nabla T$ ) and perpendicular setup ( $H \perp \nabla T$ ). The solid line shows the best fit of  $S_T^{\parallel}$  [see Eq. (3.10)] with  $\xi/D_c = 8.2 \text{ kg s}^2/\text{m}^5$  and  $\xi_{\parallel}/D_c = -1.41 \cdot 10^{-7} \text{ kg s}^2/(\text{m}^3 \text{ A}^2)$  to the experimental data ( $\square$ ). The dot-dashed line displays  $S_T^{\parallel}$  for the same parameters but  $\xi_{\parallel}/D_c = 0$ . The dashed line indicates the best fit of  $S_T^{\perp}$  [see Eq. (3.14)] with  $\xi/D_c = 8.2 \text{ kg s}^2/\text{m}^5$  and  $F = 3.75 \cdot 10^{-2} \text{ kg s}^2/(\text{m}^5 \text{ A})$  to the experimental data ( $\triangle$ ). For  $F$  and all other values see text.

Since no analytical solution for that boundary value problem is known, the following coarse approximation is made:  $(\partial c_1/\partial y)_{z=\pm h/2}$  shall be a constant  $C$  for all  $H$ -values tested here. The global Soret coefficient in the perpendicular setup can be approximated than by

$$S_T^{\perp} = \frac{h}{(T_2 - T_1) c_1 c_2} F \frac{M}{\frac{\xi}{D_c}} + \frac{S_T + \frac{\xi}{D_c} \frac{1}{c_1 c_2 \rho} \frac{\partial \mu_c^m}{\partial T}}{1 + \frac{\xi}{D_c} \frac{\partial \mu_c^m}{\partial \rho^{(1)}}}, \quad (3.14)$$

where  $F = (\xi_{\perp} C)/D_c$  will be used as the only fit parameter since  $\xi/D_c$  was determined in the parallel setup. With  $T_2 - T_1 = 1 \text{ K}$ ,  $\phi = 0.2$ , and  $h = 1 \text{ mm}$  [24], the best fit yields  $F = 3.75 \cdot 10^{-2} \text{ kg s}^2/(\text{m}^5 \text{ A})$ . The inferior match with the experimental data (see  $\triangle$  and dashed line in Fig. 2) in comparison with the parallel setup is due to the approximation that  $(\partial c_1/\partial y)_{z=\pm h/2}$  is constant. In the real system it will depend on the magnetic field since the solution for  $c_1$  depends on the magnetic field.

#### IV. CONCLUSION

The ferrofluid dynamics theory is applied to thermodiffusive problems in magnetic fluids in the presence of magnetic fields, where the MF is considered as a binary mixture. In the frame work of this theory the chemical potential could be determined analytically. Also a general expression for the mass flux is given which is independent of the fluid properties, temperature distribution and assumptions about the concentration of the nanoparticles. Applying these results to the experiments [24], their data could be interpreted better (see Fig. 2) than with the previous theory [25] which gave values about three orders of magnitude too small. Three transport coefficients, which are inherent parts the macroscopic ferrofluid dynamics theory [9], had to be used to fit this theory with the only sets of experiments available at present. In general, it is shown that for thermodiffusive problems in magnetic fluids, i.e. in colloidal suspensions sensitive to external fields, anisotropic mass fluxes are relevant and no small contributions for any nonzero strengths of the magnetic field. To elucidate this insight, more well designed experiments and further theoretical analyses are needed to improve the knowledge about thermodiffusive processes in magnetic fluids.

## V. ACKNOWLEDGEMENT

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